Spectroscopic study on ion irradiated calcites and gypsum *

N. Schöppner †[1], *S. Dedera*[1], *U.A. Glasmacher*[1], *M. Burchard*[1], *M. Zdorovets*[2,3], *and Christina Trautmann*[4]

[1]Institute of Earth Sciences, University of Heidelberg, Germany; [2]Institute of Nuclear Physics, Almaty, Kazakhstan; [3]Gumilyov Eurasian National University, Astana, Kazakhstan; [4]TU Darmstadt and GSI Helmholtzzentrum Darmstadt, Germany

Within geosciences, the use of fission tracks in various minerals as a thermochronological analytical technique is of high importance. The visualization of spontaneous and induced fission tracks uses the well established etching technique. Carbonate minerals have been tested in the past with differentiated results. The latest research clearly indicates that fission tracks develop in carbonate minerals and that they can be revealed by specific etching conditions [1], [2]. As carbonate minerals are excellent minerals for spectroscopic analytical techniques, the application of those techniques to non-destructively deduce information on fission tracks was tested.

Various carbonate minerals were irradiated within two different ion energy ranges. Ions of GeV kinetic energy were available at the GSI, while irradiations at lower energy were performed at the RGP Institute Yadernoi Fiziki, Astana, Kazakhstan. Mono crystalline samples of carbonate used in these experiments are trigonal calcite ($CaCO_3$), rhomboedric aragonite ($CaCO_3$), monocline malachite ($Cu_2[(OH)_2/CO_3]$), trigonal rhodochrosite ($MnCO_3$), trigonal dolomite ($MgCO_3$), and monocline gypsum ($CaSO_4$*$2H_2O$). At GSI, the carbonate minerals were irradiated at the UNILAC accelerator with 11.1 MeV/u ^{197}Au and ^{209}Bi ions, applying fluences of 1×10^6, 5×10^6, 1×10^7, 5×10^7, 1×10^8, 1×10^{11}, and 1×10^{12} ions/cm^2. At the RGP Institute, the samples were irradiated at the Astana accelerator with 1.7 MeV/u ^{84}Kr ions applying fluences between 1×10^{10} and 1×10^12 ions/cm^2. The lower mass and energy of these Kr ions is closer to the conditions given by natural fission fragments from ^{238}U.

All crystals were analyzed by Raman spectroscopy using the LabRam HR800 UV spectrometer equipped with an OLYMPUS BXFM-ILHS optical microscope, a grating with 1800 grooves per millimeter and a Peltier-cooled CCD detector. During the spectroscopic measurements an objective of 50x magnification was used. The spectra were excited by laser light of wavelength 473.03 nm. The lateral resolution was $\sim$2 μm, the wave number accuracy 0.5 cm^{-1} and the spectral resolution was 1 cm^{-1}. First results are provided for irradiated calcite and gypsum (Figs. 1, 2). The revealed Raman spectra are similar up to a fluences of 5×10^{11} ^{84}Kr-ions/cm^2. Above those fluences a new Raman band appears at about $\sim$430 cm^{-1}. In addition, at 5×10^{11} ^{84}Kr-ions/cm^2 a shoulder appears at the left side of the main Raman band at about 1100 cm^{-1}.

Significant changes in the Raman spectra of gypsum appear above a fluence of 1×10^{10} ^{84}Kr-ions/cm^2. With increasing fluences the peak intensities decrease and the full width at half maximum of the bands (FWHM) become larger. This applies especially to the Raman band at 3404 cm^{-1}, which characterizes the H_2O band (Fig. 1). The amplitude decrease is an indication of irradiation-induced release of water. Similar effects were observed for irradiation of gypsum with neutrons [1, 2].

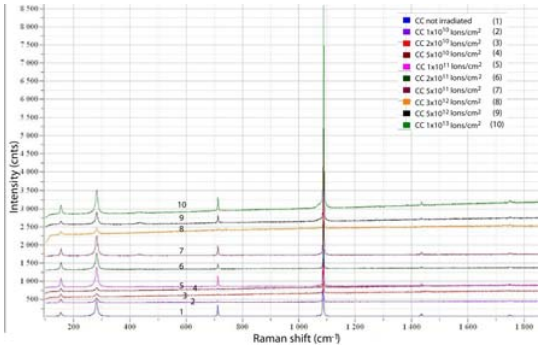

Figure 1: Spectra of calcites irradiated with fluences between 1×10^{10} and 1x10^{13} ^{84}Kr-ions/cm^2 and a spectrum of a non-irradiated calcite crystal.

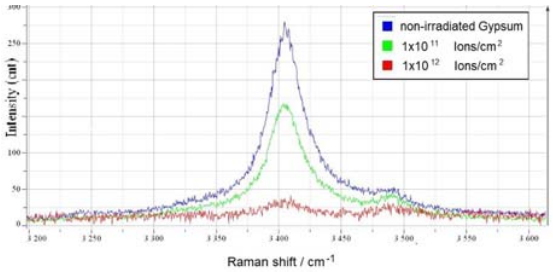

Figure 2: Raman spectra of gypsum before and after irradiation with ^{84}Kr -ions.

References

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† nicole.schoeppner@geow.uni-heidelberg.de